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EXTRACTING INFINITE SYSTEM PROPERTIES FROM FINITE SIZE CLUSTERS: "PHASE RANDOMIZATION/BOUNDARY CONDITION AVERAGING"

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ABSTRACT

When electron-electron correlations are important, it is often necessary to use "exact" numerical methods, such as Lanczös diagonalization, to study the full many-body Hamiltonian. Unfortunately, such exact diagonalization methods are restricted to small system sizes. We show that if the Hubbard U term is replaced by a "periodic Hubbard" term, the full many body Hamiltonian may be exactly solved, even for very large systems, though for low fillings. However, for half-filled systems and large U this approach is not only no longer exact, it no longer improves extrapolation to larger systems. We discuss how generalized "randomized variable averaging" (RVA) or "phase randomization" schemes can be reliably employed to improve extrapolation to large system sizes in this regime. This general approach can be combined with any many-body method and is thus of broad interest and applicability.

INTRODUCTION

Solutions of the Peierls-Hubbard Hamiltonian (PHH) [1] for small clusters are strongly dependent on the boundary condition (BC); *i.e.*, for the case of polyacetylene, whether the carbon atoms are viewed as being on a real ring, such as benzene, a real chain, such as (1,3,5)-hexatriene, or a more exotic geometry, such as anti-periodic BCs. We have found that [2,3] to reduce finite size corrections to the calculated optical gaps and spectra, thus improving the extrapolation to the infinite case, it is effective to employ novel "averaging" techniques to "randomize" the many-body levels which influence those correlation functions of interest. Briefly, by randomization, we mean simply that the total energy of the system is to be viewed as a weighted average of the energies derived separately for each of several different "random" values (RV) of a particular parameter, which may be the BC, a hopping integral, an on-site energy, or the Hubbard U: $E = \sum_{\{RV\}} x_{RV} E[RV]$, where x_{RV} is a normalized weighting factor with $\sum_{\{RV\}} x_{RV} = 1$. Similarly, any correlation function can be viewed as the average of of its values from the separate RVs. The number of RV, N_{RV}, may be varied, and the choice of the RV and x_{RV} may be truly random, or may be a pre-specified set of values, which may in turn depend on the quantity being studied. The case of *phase* randomization and its relation to Bloch's theorem are described below. More generally, this randomized-variable-averaging (RVA) technique may be viewed as a procedure to mimic disorder.

We focus in this manuscript on the calculation via exact Lanczös diagonalization of the self-consistent uniform dimerization and one-photon optical absorption of the ground state within the 1-D, 1/2-filled, one-band PHH. However, RVA is equally applicable to the calculation of, *e.g.*, the phonon modes of the ground state, as well as the geometry and self-consistent absorptions of doped or higher lying (*e.g.*, triplet) states [3], luminescence spectra, structure factors, *etc.*, in single- or multi-band models in 1-, 2-, and 3-D, via Lanczös diagonalization, as well as other numerical procedures on small lattices, such as Monte Carlo.

PHASE BOUNDARY CONDITION AVERAGING AND BLOCH'S THEOREM

Bloch's theorem tells us that the single particle wavefunctions of a system of size $M \cdot N$ with periodicity N and periodic boundary conditions (PBCs) are exactly the single particle wavefunctions of the ensemble of systems of size N for each of the phase BCs $\psi(N+1) = e^{i\phi}\psi(1)$, $\phi = 2\pi\ell/M$, $\ell = 1, ..., M$. Restated, the (exact) properties of a larger (single particle) system may be found by forming a (particular) average over smaller systems with different BCs. This "boundary condition averaging" (BCA) or "phase randomization" is a special case of our general RVA procedure. Blochs theorem may be generalized to many particle wavefunctions and is applicable here if the form of the Hamiltonian is modified slightly. The one-dimensional PHH is

$$H = \sum_{\ell,\sigma}(-t_0 + \alpha\delta_\ell)(c^\dagger_{\ell\,\sigma}c_{\ell+1\,\sigma} + c^\dagger_{\ell+1\,\sigma}c_{\ell\,\sigma}) + U\sum_\ell n_{\ell\uparrow}n_{\ell\downarrow} + \frac{1}{2}K\sum_\ell \delta_\ell^2 \qquad (1)$$

If we replace the Hubbard term, $U\sum_{\ell=1}^{N\cdot M} n_{\ell,\uparrow}n_{\ell,\downarrow}$, by its periodic analog,

$$H_{el-el} \rightarrow \frac{U}{M}\sum_{\mu,\nu=1}^{M}\sum_{\ell=1}^{N} n_{\ell+\mu\cdot N,\uparrow}n_{\ell+\nu\cdot N,\downarrow}\ , \qquad (2)$$

then, if the original problem had periodicity N, $\langle n_{\ell,\sigma}\rangle = \langle n_{\ell+\nu\cdot N,\sigma}\rangle$, we have, in a mean-field sense, the "same" Hamiltonian. With this new "periodic-Hubbard" Hamiltonian (periodic PHH) the Bloch analysis of a given large (periodic) system ($N\cdot M$ sites, $N_e\cdot M$ electrons) is accomplished by considering a small system (N sites, N_e electrons) with several different BCs. The method scales linearly with M, allowing one to handle reasonably large systems, and sets a sound theoretical basis for the empirical observation [2] that RVA can be used to smooth optical absorption spectra obtained via exact finite-size diagonalization.

Using the periodic-Hubbard term, Eq. (2), assuming the lattice distortion has the same periodicity, $\delta(\ell+N) = \delta(\ell)$, and using $\sum_{\mu=1}^{M}\sum_{\ell=1}^{N}\sum_{\sigma=\uparrow,\downarrow} n_{\ell+\mu\cdot N,\sigma} \equiv N_e\cdot M$, one can show:

$$H_{per} = \frac{1}{M}\sum_{\mu,\nu=1}^{M}\sum_{i,j,k,l=1}^{N}\sum_{s_1,s_2=\uparrow,\downarrow} c^\dagger_{i+\mu\cdot N,s_1}c_{j+\mu\cdot N,s_1} \times H(i,j,k,l;s_1,s_2)\, c^\dagger_{k+\nu\cdot N,s_2}c_{l+\nu\cdot N,s_2} \qquad (3)$$

where

$$\begin{aligned} H(i,j,k,l;s_1,s_2) &= H(i+N,j+N,k,l;s_1,s_2) \\ &= H(i,j,k,l;s_2,s_1) = H(k,l,i,j;s_1,s_2) = H(j,i,k,l;s_1,s_2) \end{aligned}$$

and

$$\begin{aligned} H(i,j,k,l;s_1,s_2) &= \frac{1}{2}\left[\sum_{n=1}^{N}(-t_0+\alpha\delta_n)(\partial_{i,n}\partial_{j,n+1}+\partial_{i,n+1}\partial_{j,n})\frac{\partial_{k,l}}{N_e} + (i,j \rightleftharpoons k,l)\right] \\ &+ \frac{1}{2}K\sum_{n=1}^{N}\delta_n^2\frac{\partial_{i,j}}{N_e}\frac{\partial_{k,l}}{N_e} + U\partial_{s_1,s_2}\partial_{i,j}\partial_{k,l}\ . \end{aligned}$$

Note that $H(i,j,k,l;s_1,s_2)$ is independent of μ, ν, and M. The Bloch analysis on the many-body eigenfunctions using the symmetries of $H(i,j,k,l;s_1,s_2)$ leds to a Ψ of the form:

$$\Psi_{k_1,\ldots,k_p} = \sum_{\sigma_1,\ldots,\sigma_p=\uparrow,\downarrow} \sum_{n_1,\ldots,n_p=1}^{N} \psi_{k_1,\ldots,k_p}(n_1,\ldots,n_p) \times \sum_{\mu_1=1}^{M} c^{\dagger}_{n_1+\mu_1\cdot N,\sigma_1} e^{ik_1\mu_1} \times \cdots \times \sum_{\mu_p=1}^{M} c^{\dagger}_{n_p+\mu_p\cdot N,\sigma_p} e^{ik_p\mu_p} \tag{4}$$

where $\psi_{k_1,\ldots,k_\ell,\ldots,k_p}(n_1,\ldots,n_\ell,\ldots,n_p) = \psi_{k_1,\ldots,k_\ell,\ldots,k_p}(n_1,\ldots,n_\ell+\mu_\ell\cdot N,\ldots,n_p)$, $k_\ell = (2\pi j/M)$, $j\epsilon\{0,\ldots,M-1\}$, and $p \equiv N_e\cdot M$. We find for $i,j\epsilon\{1,\ldots,N\}$,

$$\sum_{\nu_1=1}^{M} c^{\dagger}_{i+\nu\cdot N,s} c_{j+(\nu+\nu_0)\cdot N,s} \Psi_{k_1,\ldots,k_p} = \sum_{\sigma_1,\ldots,\sigma_p=\uparrow,\downarrow} \sum_{n_1,\ldots,n_p=1}^{N} \sum_{\ell=1}^{N} \psi_{k_1,\ldots,k_p}(n_1,\ldots,n_p)\, \partial_{n_\ell,j}\partial_{\sigma_\ell,s}\, e^{ik_\ell\nu_0} \times \sum_{\mu_1=1}^{M} c^{\dagger}_{n_1+\mu_1\cdot N,\sigma_1} e^{ik_1\mu_1} \times \cdots \times \sum_{\mu_\ell=1}^{M} c^{\dagger}_{i+\mu_\ell\cdot N,\sigma_\ell} e^{ik_\ell\mu_\ell} \times \cdots \times \sum_{\mu_p=1}^{M} c^{\dagger}_{n_p+\mu_p\cdot N,\sigma_p} e^{ik_p\mu_p} \tag{5}$$

(ν_0 comes into play for operators like $c^{\dagger}_{N,s}c_{N+1,s}$). From Eq. (5) we see H_{per} is diagonal in the k's,

$$H_{per}\ \Psi^{\beta}_{k_1,\ldots,k_p} = E_\beta\ \Psi^{\beta}_{k_1,\ldots,k_p}\ , \tag{6}$$

as are $\rho = \sum_{\ell,\sigma} n_{\ell,\sigma}$ and $J = i\sum_{\ell,\sigma}(-t_0+\alpha\delta_\ell)(c^{\dagger}_{\ell,\sigma}c_{\ell+1,\sigma} - c^{\dagger}_{\ell+1,\sigma}c_{\ell,\sigma})$, and the eigenfunctions of H_{per} can be written in the form Eq. (4).

The symmetry of Ψ implies that if $\sigma_{\ell_1} = \sigma_{\ell_2}$ and $k_{\ell_1} = k_{\ell_2}$ then $n_{\ell_1} \neq n_{\ell_2}$. Thus there are at most $2N$ of the k_ℓ the same. We postulate that the ground state lies in the manifold with each of the M distinct values for k_ℓ occurring N_e times and (for the half-filled band) with equal numbers of up and down spins (in general, the largest manifold). This can be checked, and we stress that the analysis up to this point is exact for the periodic PHH. In this manifold we can write Ψ as:

$$\Psi^{\beta} = \sum_{\alpha_1,\ldots,\alpha_M=1}^{2N} \phi^{\beta}(\alpha_1,\ldots,\alpha_M)\ \Psi^{N_e}_{q_1,\alpha_1} \cdots \Psi^{N_e}_{q_M,\alpha_M}\ , \tag{7}$$

where $q_\ell \equiv (2\pi\ell/M)$ and

$$\Psi^{N_e}_{q,\alpha} = \sum_{\sigma_1,\ldots,\sigma_{N_\uparrow}=\uparrow} \sum_{\sigma_{N_\uparrow+1},\ldots,\sigma_{N_e}=\downarrow} \sum_{n_1,\ldots,n_{N_e}=1}^{N} \psi^{N_e}_{q,\alpha}(n_1,\ldots,n_{N_e}) \times \sum_{\mu_1=1}^{M} c^{\dagger}_{n_1+\mu_1\cdot N,\sigma_1} e^{iq_1\mu_1} \times \cdots \times \sum_{\mu_{N_e}=1}^{M} c^{\dagger}_{n_{N_e}+\mu_{N_e}\cdot N,\sigma_{N_e}} e^{iq_{N_e}\mu_{N_e}}$$

where $N_\uparrow = [[\frac{(N_e+1)}{2}]]$, $N_\downarrow = [[\frac{N_e}{2}]]$ ([[ ]] denotes integer value of). Note

$$c^{\dagger}_{1+N,\sigma}c_{N,\sigma}\ \Psi^{N_e}_{q,\alpha} = e^{iq}\ c^{\dagger}_{1,\sigma}c_{N,\sigma}\ \Psi^{N_e}_{q,\alpha}\ , \tag{8}$$

and so solving the N_e electron problem on the full $N\cdot M$ sites with PBCs for fixed q is *exactly* equivalent to solving

$$H_{per}\ \Psi^{N_e}_{q,\alpha} = E^{N_e}_{q,\alpha}\ \Psi^{N_e}_{q,\alpha} \tag{9}$$

on N sites with the q-dependent BC defined by Eq. (8). Thus, to find the exact N_e electron eigenfunctions $\Psi^{N_e}_{q,\alpha}$ on $N\cdot M$ sites, we only need to solve Eq. (9) with N sites and N_e electrons for each of the M BCs, rather than the full $N\cdot M$ site problem.

To obtain the exact eigenfunctions of the full $N_e \cdot M$ electron problem, we need to solve for $\phi^\beta(\alpha_1, ..., \alpha_M)$. This is where an approximation must be made to be able to solve the problem numerically, as we assume N is already as large as computationally feasible. The full eigenvalue problem yields

$$\left[H_{per} \ , \ \Psi^{N_e}_{q,\alpha} \right] = E^{N_e}_{q,\alpha} + \frac{U}{M} \sum_{\ell=1}^{N} \left\{ \left[\sum_{\mu=1}^{M} n_{\ell+\mu\cdot N,\uparrow} \ , \ \Psi^{N_e}_{q,\alpha} \right] \sum_{\nu=1}^{M} n_{\ell+\nu\cdot N,\downarrow} + \left[\sum_{\nu=1}^{M} n_{\ell+\nu\cdot N,\downarrow} \ , \ \Psi^{N_e}_{q,\alpha} \right] \sum_{\mu=1}^{M} n_{\ell+\mu\cdot N,\uparrow} \right\} \tag{10}$$

To solve Eq. (6) we must use some approximation for Eq. (10), such as perturbation theory or mean-field. Thus BCA means we treat electron-electron correlations within the N_e electron manifold exactly, and electron-electron correlations between N_e electron manifolds approximately. The BCA results reported here are zero-order perturbation theory: we have assumed $\phi^\beta(\alpha_1, ..., \alpha_M)$ to be a product of δ-functions; *i.e.*, for the ground state we use:

$$\Psi^0 \simeq \Psi^{N_e}_{q_1,0} \cdots \Psi^{N_e}_{q_M,0} \ ; \qquad E_0 \simeq \sum_{\ell=1}^{M} E^{N_e}_{q_\ell,0} \ . \tag{11}$$

Note that if we had been interested in systems at low filling (N_e electrons), rather than near half filling ($N_e \cdot M$ electrons), there would be no need to make any approximations.

Comparison of the periodic and standard Peierls-Hubbard Models at half-filling

At U=0 Bloch's theorem for the many particle wavefunctions as formulated above is exact at all fillings. For the half-filled PHH, we have just shown that phase BCA involves two approximations: first, replacing the Hubbard term by Eq. (2), and, second, ignoring correlations between the N_e particle wavefunction leading to Eq. (11). We now test these approximations. In Fig. 1 we show the minimum energy dimerization of an 8-site system as a function of U for (*i*) phase averaged solution and (*ii*) the exact solution of the half-filled band with the periodic Hubbard term and periodicity 2,4, and 8 (periodicity 8 is the the usual Hubbard). The approximate periodic (BCA) solution lies between the exact periodic solution and the exact pure-Hubbard solution. Since the original problem was for $U \cdot M \sum_{\ell=1}^{N\cdot M} n_{\ell\uparrow} n_{\ell\downarrow}$ on $N \cdot M$ sites, one might argue that the approximate phase-averaged result for $U \sum_{\ell=1}^{N} n_{\ell\uparrow} n_{\ell\downarrow}$ on N sites may be closer to the desired answer than the exact periodic-Hubbard $N \cdot M$ site result.

In Fig. 2 we show the dimerization amplitude obtained by phase BCA. We see that the extrapolated infinite behavior at small to intermediate U is well approximated after phase averaging for N=10 even without extrapolation. The agreement is best for small U, while no change from the pure periodic behavior is found for large U. That this particular technique is expected to have no effect at large U can be seen by examining the effective spin-Peierls Hamiltonian [4], which is asymptotically independent of the phase of the BC. However, the phase-averaged analysis does lead to the interpretation of the finite-size result as the result for the larger system with an effective periodic-Hubbard interaction. Thus, if U is scaled by the system size, as is done in Fig. 2, the infinite behavior should be more easily extrapolated. It appears that this is at least approximately true.

To show how phase BCA affects the optical spectrum, in Fig. 3a we show the (Lorentzian broadened) spectrum at U/t_0=0.4 for N=8 and 5 phase BCs (enough to yield a smooth spectrum at U=0). It is clear how the different BCs "fill in" the spectrum. Fig. 3b shows the spectrum at U/t_0=1.6. Here, despite the several BCs used, the spectrum remains sparse, due to the loss of effectiveness of the phase BCA scheme as discussed above. If we were to use a broad enough Lorentzian to smooth this spectrum

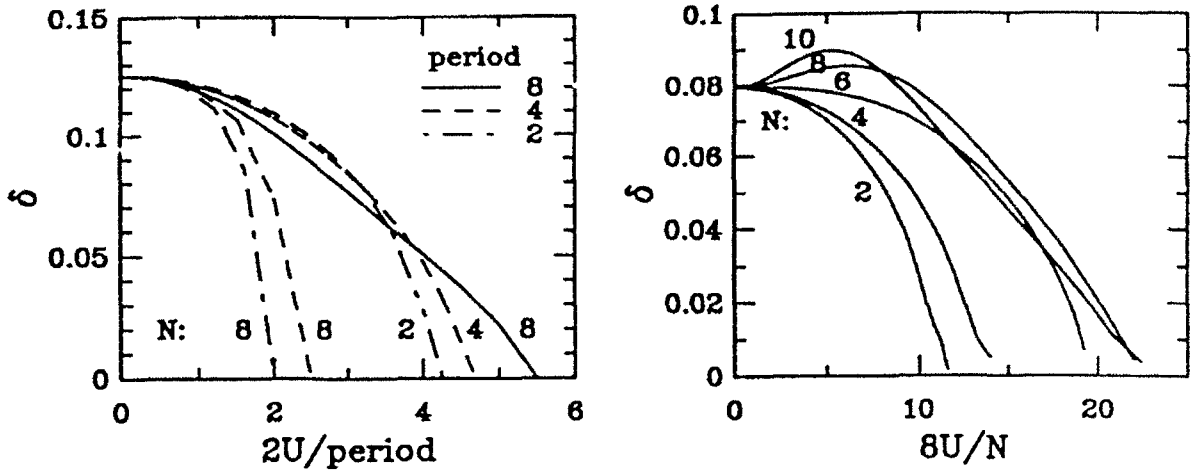

Fig. 1 (left). Comparison of the approximate and exact periodic-Hubbard minimum energy dimerization as a function of U for t_0=2.5 eV, α=4.1 eV/Å, and K=21 eV/Å^2.

Fig. 2 (right). Dimerization *vs.* U for various N: phase BCA. Parameters as Fig. 1.

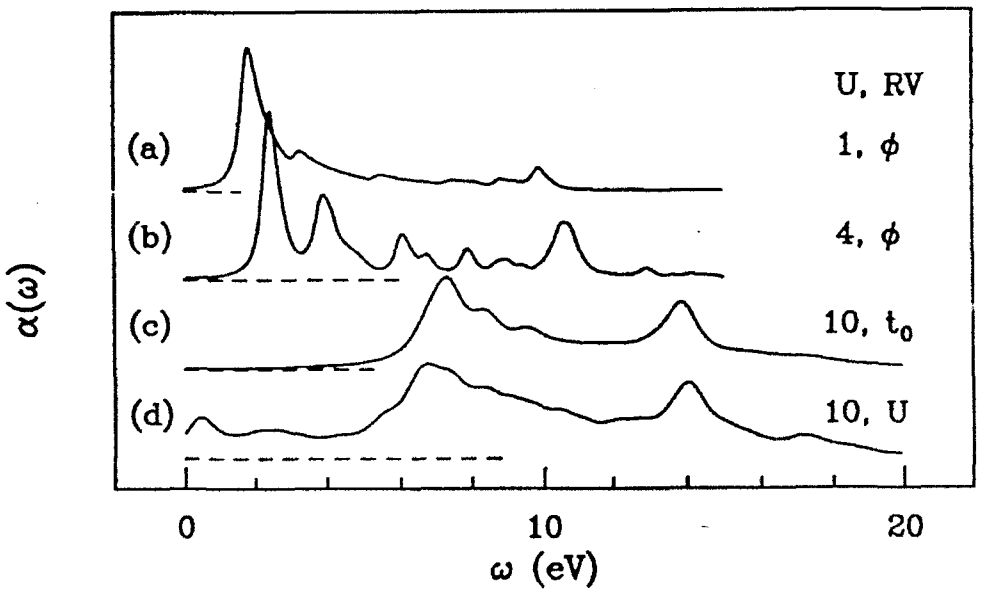

Fig. 3. The Lorentzian broadened optical absorption spectra calculated using the phase BCA method for (a) U=1 eV and (b) U=4 eV. Note the "sparseness" of the spectrum in the latter case indicating the failure of the phase BCA approach. For comparison, we also show the spectrum at U=10 eV using (c) amplitude BCA and (d) an RVA procedure where U is varied on one site. Note these RVA spectra are dense and show the same features. Parameters as Fig. 1 but δ=0.14 Å.

AMPLITUDE BOUNDARY CONDITION AVERAGING

The modification of the phase of the BC discussed above can also be viewed as passing a random magnetic flux through an ensemble of closed ring and studying average properties. In this sense we have "randomized" the locations of the momentum space states. We have already indicated that RVA can help us transcend the limitations of this "bond phase/magnetic flux" approach. We can "randomize" electronic properties by, *e.g.*, changing a local hopping or an on-site energy or a Coulomb repulsion somewhere on the chain. One could also introduce an additional field and vary it about zero. Although

[5], there is as yet no provably accurate prescription for for arbitrary U and V. However, certain intuitive rules must guide us. First, whatever change is made to randomize must, of course, do so effectively. Second, the change in the system must be negligible as the lattice size is increased to infinity; for example, if only one bond or site is varied from calculation to calculation, then the effect of such a change is immaterial in the thermodynamic limit. Finally, the behavior for small lattice sizes must be illustrative of the infinite-size limit. Put another way, one must still be able to make a reasonable extrapolation to the infinite chain.

We have found [2] an "amplitude BCA" ("scaled-hopping") technique to be effective and produce results in good agreement with expectations based on both strong- and weak-coupling analytic arguments. For this method, we randomize by varying the *magnitude* rather than the phase of the "boundary" hopping – *i.e.*, between sites 1 and N – typically from $-t_0$ to $+t_0$ in ten to twenty equal steps. The individual spectra are then added together with weights x_i chosen to minimize the total length of the final curve, though giving each spectra equal weight yields virtually identical results. This clearly incorporates the special case of using only periodic (x=1) or antiperiodic (x=−1) rings, the JT/nJT difference being important for weak coupling. It also incorporates the case of the open chain (x=0), which has the "best" single BC size dependence (though with attendant "end effect" problems). Finally, it works in the strong-coupling limit.

In Fig. 3c, we show the spectra produced by the amplitude BCA technique for for a larger U than that in Fig. 3b where phase BCA failed. In Ref. [2] we showed that not only are the gross features of the spectra obtained by amplitude BCA in agreement with strong-coupling calculations [6], but in addition they show substantially more interesting detail, such as the "decoupled-dimer" peak located at $\sim U/2 + \sqrt{(U/2)^2 + (2t_0)^2}$ in systems with strong electron-phonon *and* electron-electron couplings. To emphasize the generality, in Fig. 3d we show the spectrum obtained by varying U on the first site between 0 and 20 eV. This large U variation produces states in the optical gap, but otherwise the spectrum is unchanged from Fig. 3c. We feel that these results, and those in Ref.s [2,3], demonstrate that RVA is an effective means for reducing (and in some cases practically eliminating) finite size dependence, yielding results that can be confidently extrapolated to the infinite size limit, and which are in good agreement with known analytic results.

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